

# Study of the superconducting and magnetic properties of niobium doped $\text{RuSr}_2\text{Gd}_{1.5}\text{Ce}_{0.5}\text{Cu}_2\text{O}_{10-\delta}$ ruthenocuprates

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## Abstract

Polycrystalline samples of  $\text{Ru}_{1-x}\text{Nb}_x\text{Sr}_2\text{Gd}_{1.5}\text{Ce}_{0.5}\text{Cu}_2\text{O}_{10-\delta}$ ,  $0 \leq x \leq 0.5$ , have been synthesized and studied by resistivity, magnetization and ac susceptibility measurements. It was identified a strong suppression of the spin glass (SG) transition, which was totally suppressed in samples for  $x \geq 0.2$ . The hysteresis loops at low temperatures are the result of the contribution of two distinct magnetic phases: a canted AFM phase and the SG phase. More importantly, the significant changes in the magnetic response of the material affect the superconducting properties of the samples. It was found that both  $T_c$  and the superconducting fraction are reduced in samples that show the spin glass phase, possibly due to the magnetic pair breaking effect.

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**Keywords:** Ruthenocuprate; Magnetic order; Magnetism-superconductivity coexistence; Spin glass; Substitution effect; Niobium doping

## 1. Introduction

The coexistence of superconductivity and magnetism at a microscopic level in the ruthenocuprate family  $\text{RuSr}_2\text{Gd}_{1.5}\text{Ce}_{0.5}\text{Cu}_2\text{O}_{10-\delta}$  (Ru-1222) is still a controversial subject. One strategy to explore this topic is by careful chemical substitution of the parent composition. The fact that superconductivity occurs in the  $\text{CuO}_2$  planes, while the magnetic order is restricted to the  $\text{RuO}_2$  planes, permits to chemically modify one of these families of planes keeping the other one unaffected. The niobium substitution for Ru, in particular, is very attractive, since in this case both ions have valence close to  $5^+$  and changes in the carrier density should be very small. Therefore, it is expected that changes in the superconductivity will be mostly due to the possible coupling of magnetism and superconductivity in this system.

## 2. Results and discussion

Samples of composition  $\text{Ru}_{1-x}\text{Nb}_x\text{Sr}_2\text{Gd}_{1.5}\text{Ce}_{0.5}\text{Cu}_2\text{O}_{10-\delta}$ ,  $0 \leq x \leq 0.5$ , have been studied by resistivity and magnetization measurements. In Fig. 1 we present the zero field cooled (ZFC) and field cooled (FC) magnetization curves. For the sake of clarity, the measurements were divided in two groups, one for the lightly doped samples ( $x \leq 0.2$ , Fig. 1a) and a second one for the heavily doped samples ( $0.3 \leq x \leq 0.5$ , Fig. 1b). The peak in the ZFC curve associated with the spin glass transition [1] is strongly suppressed with doping, disappearing completely for samples with  $x \geq 0.2$ . There is a dramatic reduction in the magnetic moment associated with the Ru sublattice making the superconducting signal more prominent. Curiously, the superconducting diamagnetic signal becomes weaker with the increase of niobium content, for  $x \geq 0.2$ , indicating a smaller superconducting fraction.

The suppression of the spin glass phase also affects the magnetic hysteresis loop at low temperatures (not shown). As shown in Fig. 2, all parameters which characterize a

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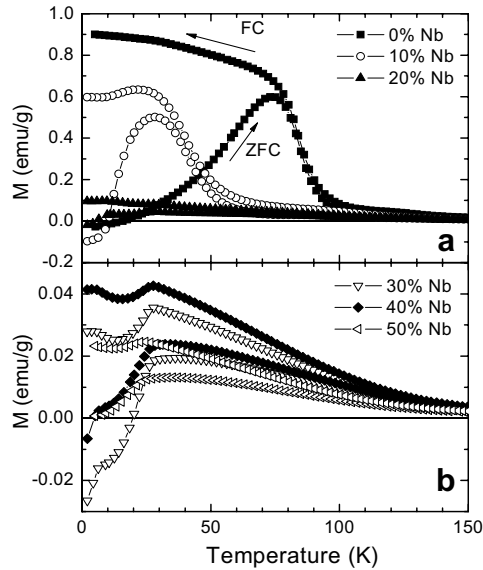


Fig. 1. DC magnetization as a function of temperature for lightly: (a) and heavily and (b) doped samples, for  $H = 50$  Oe.

hysteresis loop (coercive field  $H_c$ , remanence  $M_r$ , and saturation magnetization  $M_s$ ) present a clear change when the  $x = 0.2$  doping level is crossed. This indicates the magnetic loops have contributions from two different phases: the spin glass phase and the canted antiferromagnetic phase.

Even more interesting, the superconducting transition temperature ( $T_c$ ) extracted from resistivity measurements (not shown), also present a maximum value for  $x = 0.2$ , see Fig. 3. For samples with  $x \leq 0.2$  the larger magnetic moment associated with the spin glass phase breaks the Cooper pairs, reducing  $T_c$ . On the other hand, the reduction in  $T_c$  by increasing  $x$  is possibly due to the small differ-

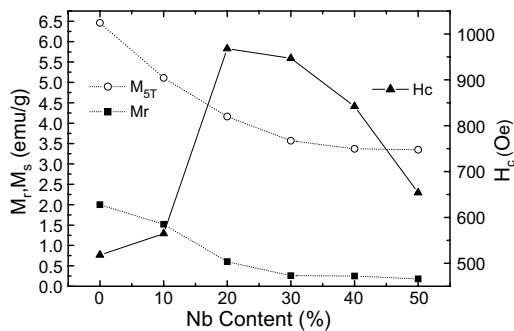


Fig. 2. Dependence of the coercive field ( $H_c$ , right axis), remanent magnetization and saturation magnetization ( $M_r$  and  $M_s$ , respectively, left axis) with niobium doping.

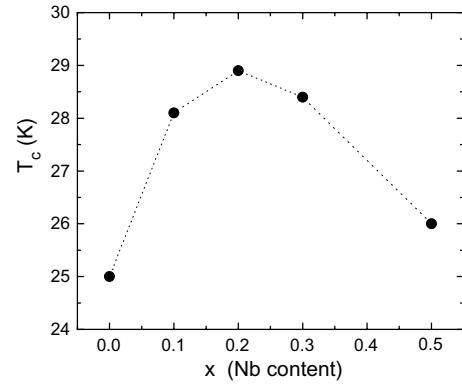


Fig. 3. Superconducting transition temperature, extracted from resistivity measurements, as a function of niobium doping.

ence in the valence from Ru to Nb atoms, which reduces the carrier density, thus reducing  $T_c$  and the superconducting fraction as a consequence.

### 3. Conclusions

In this work we report that niobium substitution strongly reduces the spin glass transition temperature, which is completely suppressed for  $x \geq 0.2$ . The non-monotonic behavior of  $T_c$  is explained through a decrease in the carrier density, caused by an increase of Nb doping, combined with a pair breaking effect of the strong magnetic moment from the SG phase, present in the samples with  $x = 0.0$  and  $x = 0.1$ . Therefore, we verified that the magnetic moment does interfere in the superconducting properties of Ru-1222:Nb samples, in contrast to previous studies which indicated both states to be decoupled [2,3]. Also, the magnetic hysteresis loops provide additional evidence that the magnetic behavior of the Ru-1222:Nb samples may be interpreted as a combination of two separate contributions, reinforcing the magnetic phase separation scenario.

### Acknowledgement

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### References

- [1] C.A. Cardoso, F.M. Araujo-Moreira, V.P.S. Awana, E. Takayama-Muromachi, O.F. de Lima, H. Yamauchi, M. Karppinen, Phys. Rev. B 67 (2003) 020407.
- [2] H.K. Lee, Y.C. Kim, Int. J. Mod. Phys. B 17 (2003) 3682.
- [3] H.K. Lee, G.V.M. Williams, Physica C 415 (2004) 172.